

# Coherence-incoherence crossover in the normal state of iron-oxypnictides and importance of the Hund's rule coupling

K. Haule and G. Kotliar

Department of Physics, Rutgers University, Piscataway, NJ 08854, USA

E-mail: haule@physics.rutgers.edu

PACS numbers: 71.27.+a, 71.30.+h

**Abstract.** A new class of high temperature superconductors based on iron and arsenic was recently discovered [1], with superconducting transition temperature as high as 55 K [2]. Here we show, using microscopic theory, that the normal state of the iron pnictides at high temperatures is highly anomalous, displaying a very enhanced magnetic susceptibility and a linear temperature dependence of the resistivity. Below a coherence scale  $T^*$ , the resistivity sharply drops and susceptibility crosses over to Pauli-like temperature dependence. Remarkably, the coherence-incoherence crossover temperature is a very strong function of the strength of the Hund's rule coupling  $J_{Hund}$ . On the basis of the normal state properties, we estimate  $J_{Hund}$  to be  $\sim 0.35$  eV. In the atomic limit, this value of  $J_{Hund}$  leads to the critical ratio of the exchange constants  $J_1/J_2 \sim 2$ . While normal state incoherence is in common to all strongly correlated superconductors, the mechanism for emergence of the incoherent state in iron-oxypnictides, is unique due to its multiorbital electronic structure.

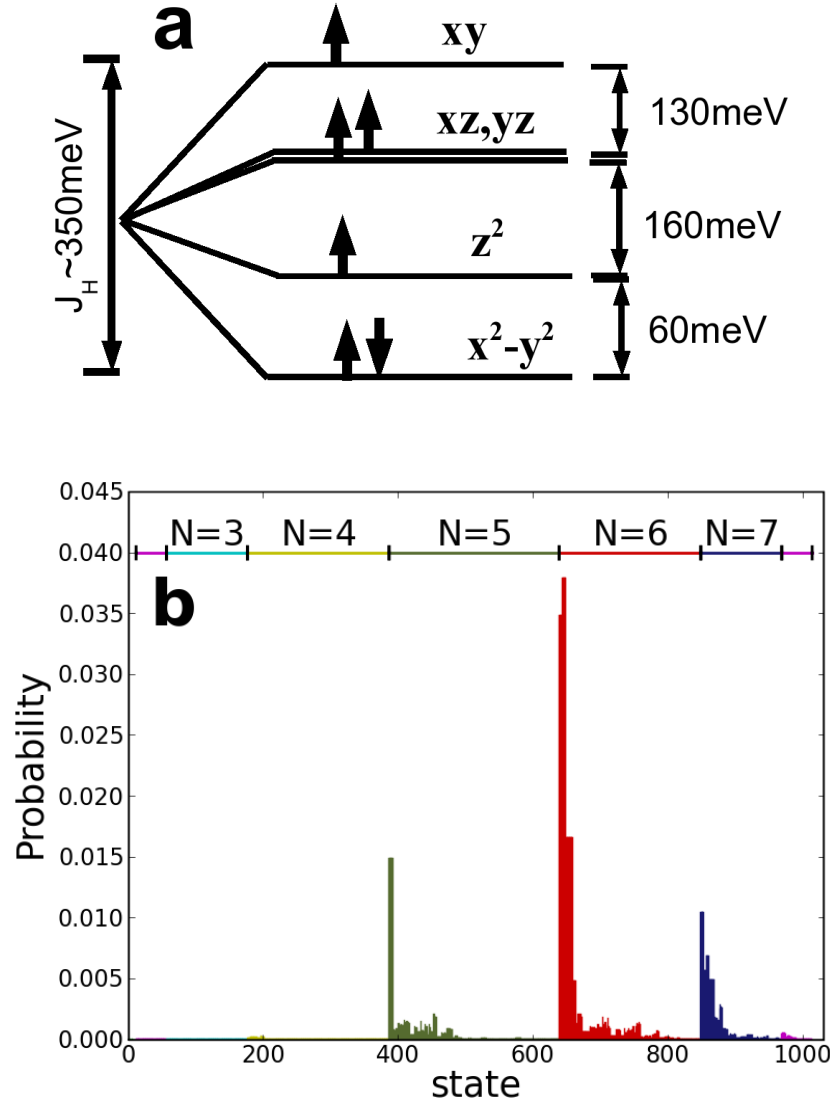
The unusually high superconducting critical temperatures in iron-oxypnictides together with unusual normal state properties, which do not fit within the standard framework of the Fermi liquid theory of solids, place the iron pnictides in the broad category of strongly correlated superconductors, such as  $\kappa$  organics, cerium and plutonium based heavy fermions, and cuprate high temperature superconductors. In all these materials, superconductivity emerges in close proximity to an incoherent state with unconventional spin dynamics, that cannot be describe in terms of weakly interacting quasiparticles. Describing the normal state of these materials, is one of the grand challenges in condensed matter theory and has resulted in numerous controversies in the context of the cuprates.

Iron pnictides show very high resistivity and very large uniform susceptibility in the normal state [1]. Superconductivity emerges from a state of matter with highly enhanced Pauli like type of susceptibility [3]. As a function of  $F^-$  doping, the susceptibility is peaked around 5% doping reaching a value 25 times bigger than Pauli susceptibility given by LDA [3, 4]. In the parent compound, the resistivity exhibits a peak at 150 K [1] followed by a sharp drop, which is due to a structural transition from tetragonal (space group  $P4/nmm$ ) to orthorhombic structure (space group  $Cmma$ ) [7, 3] followed by a spin density wave transition [5, 6] at lower temperature. Only a 5% doping completely suppress the specific heat anomaly [8], while resistivity still shows a very steep drop below a characteristic temperature  $T^*$  [8, 9]. While there are suggestions that the drop of resistivity is due to opening of a pseudogap and proximity to a quantum critical point in samarium compound [9], other measurements in lanthanum compound seem to suggest less exotic and more Fermi liquid-like resistivity at 10% doping level[10, 11]. Here we will show that the steep drop of resistivity in doped compounds can be understood on the basis of incoherence-coherence crossover, which is naturally accompanied by gradual screening of the magnetic moments, leading to enhanced magnetic susceptibility, and crossover from Currie-Weiss to Pauli type susceptibility.

Theoretically, electron-phonon coupling is not sufficient to explain superconductivity in  $LaO_{1-x}F_xFeAs$  [12, 13] and spin and orbital fluctuations need to be considered. The weak coupling approach based on RPA approximation has been carried out by several groups for a simplified two band model containing two orbitals,  $d_{xz}$  and  $d_{yz}$  [14, 15, 16]. The nature of the superconducting state and the pairing symmetry is apparently very sensitively to the parameters of the model, and therefore it is very desirable to reliably determine parameters of a microscopic Hamiltonian, which is the purpose of this article.

The electronic structure of the  $LaO_{1-x}F_xFeAs$  compound was studied by the density functional theory (DFT) [4] and the dynamical mean field theory (DMFT) [12]. DFT calculations show that this material has a well separated set of bands with mostly  $d$  character in the interval between  $-2$  eV and  $2$  eV around the Fermi level [12]. The parent compound has five Fermi surface sheets [4], two electron cylinders, centered around the zone edge (M-A line), and three hole pockets around the zone center ( $\Gamma$ -Z line). Upon doping with electrons, the hole-like pocket quickly disappear. The nesting wave vector

between the hole and electron pocket promotes the spin density wave instability which was actually observed in experiment [6]. Notice however that the Bragg peak in neutron experiments remains commensurate and does not change with doping, pointing to the inadequacy of the weak coupling spin density wave scenario.



**Figure 1.** a) Sketch of the orbital levels in the downfolded LDA Hamiltonian for LaOFeAs, keeping five d-orbitals (see appendix Appendix A). The levels are obtained by  $h_\alpha = \sum_{\mathbf{k}} H_{\mathbf{k},\alpha\alpha}^{LDA}$ , where  $\alpha$  is the orbital index. The Hund's coupling in this compound becomes relevant when its strength is comparable to the total splitting  $J \sim 350$  meV. The coordinate system is chosen such that  $x$  and  $y$  axis point from Fe atom towards its the nearest neighbor Fe atoms. b) Probability for each iron 3d atomic state in DMFT calculation for LaOFeAs at  $T = 116$  K and  $J_{Hund} = 0.4$  eV. There are 1024 atomic states in Fe-3d shell. Within sector with constant occupancy, states are sorted by increasing atomic energy).

Fig. 1 sketches the crystal field levels, as obtained by the Local Density

Approximation (LDA), when the localized Wannier orbitals are constructed for Fe-3d states. Notice that the tetragonal splittings are comparable to the t2g-eg splittings, leaving only two degenerate orbitals, i.e.,  $xz$  and  $yz$ . Notice however, that the bandwidth of these orbitals, as shown in Fig.3 of Ref. [12], is of the order of 3 eV, which is one order of magnitude larger than the crystal field splittings.

A naive atomic picture suggests a large magnetic moment on the Fe atom ( $S = 2$ ), and consequently almost classical moments, which can not condense by Cooper pairing, but would rather order magnetically, just like they do in pure iron. Due to degeneracy of  $xz$  and  $yz$  orbital, even an infinitesimal Hund's rule coupling would therefore lead to a spin state of at least  $S = 1$ .

Dynamical mean field studies [12], showed that the correlations in the parent compound are strong enough to make this material a bad metal with a large electronic scattering rate at, and below room temperature, as seen in the momentum resolved photoemission spectra, and the absence of a Drude peak in the optical conductivity. However, it was argued in Ref. [12], that the correlations are *not* strong enough to open the Mott gap in the parent compound, in contrast to the hole doped cuprates, which are known to be doped Mott insulators. The DMFT study determined LaOFeAs, to be a strongly correlated metal, which is not well described by either atomic physics, nor band theory. Furthermore, it was shown that all the five iron  $d$  orbitals have appreciable one particle density of states within 0.5 eV of the Fermi level, and therefore a realistic minimal model for these materials should contain all five  $d$  iron orbitals.

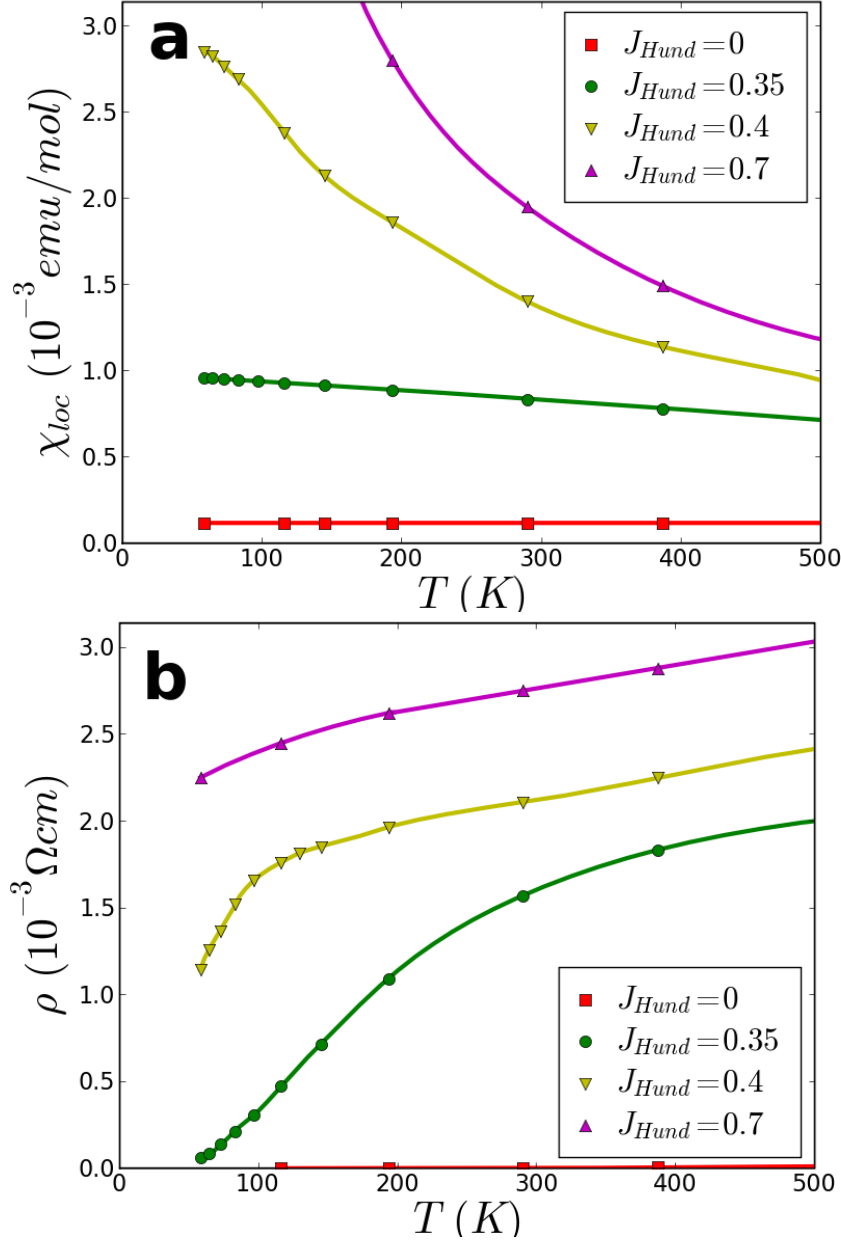
In this article, we use the LDA+DMFT method [17] in combination with the continuous time Quantum Monte Carlo method [18], to investigate the transport and thermodynamic properties of the 10% doped  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$  compound. More details of the method are given in Ref. [12, 17].

Our goal is to understand why a material with high orbital degeneracy, which naively would have expected to be an excellent metal [19], exhibits strong correlation effects, and what determines the crossover from the incoherent regime to the coherent Fermi liquid state. We show that the Hund's rule coupling  $J_{\text{Hund}}$  dramatically reduces the coherence scale  $T^*$  in these compounds, and promotes the highly incoherent metallic state.

In physical terms, correlated quasiparticles are spin one half objects that have to be built from atomic states, which are dominantly  $S = 2$  and  $S = 1$ , in the presence of Hund's rule coupling. As a result, the overlap of the quasiparticle and the bare electronic states is very small.

To understand the nature of the metallic incoherent state, it is useful to study the probability that an electron in this compound is found in any of the atomic states of iron 3d orbital. This is plotted in the atomic histogram in Fig. 1 introduced in Ref. [20]. Even the most probable atomic states have probability of only a few percent, hence a naive atomic limit is qualitatively wrong for this compound. The atomic histogram, characterizes the nature of the ground state of the material. To investigate the coherence incoherence crossover, we need to probe the excitation spectra, through the transport

*Coherence-incoherence crossover in the normal state of iron-oxypnictides and importance of the Hund's rule*  
and the thermodynamic probes.



**Figure 2.** a) The local susceptibility versus temperature for few different Hund's coupling strengths. b) Resistivity versus temperature for the same Hund's coupling strengths.

We first turn to the magnetic susceptibility which is the most direct probe of the spin response:

$$\chi = \frac{(g\mu_B)^2 N_A}{k_B} \int_0^\beta d\tau \langle S_z(\tau) S_z(0) \rangle. \quad (1)$$

Here  $S_z$  is the spin of the iron atom,  $g\mu_B/2$  is a magnetic moment of a free electron,  $N_A$  is the Avogadro number, and  $\beta$  is the inverse temperature. In the absence of screening

(static  $S_z$ ), susceptibility should follow the Curie-Weiss law  $\chi = \frac{(g\mu_B)^2 N_A S(S+1)}{3k_B T}$ . In the opposite itinerant limit, the spin susceptibility exhibits Pauli response  $\chi = \mu_B^2 N_A D_0$ , where  $D_0$  is the density of state at zero frequency. In Fig. 2a, we show the local spin susceptibility for a few values of Hund's coupling  $J_{Hund}$  in 10% doped compound.

In the absence of  $J_{Hund}$ , the susceptibility obeys the Pauli law, and the mass enhancement due to correlations is negligible. On the other hand, at large  $J_{Hund} = 0.7$  eV, the system remains in the local moment regime down to lowest temperature we reached (50 K). For some intermediate values of Hund's coupling  $J_{Hund} \sim 0.35 - 0.4$  eV, the system evolves from an incoherent metal at high temperature, to the coherent Fermi liquid at low temperature.

The metallicity of the compound always leads to screening of the electron magnetic moments, which eventually disappear at sufficiently low temperature and frequencies, but are present at high temperature and high energies. The scale at which the screening of the local moment takes place, however, is a very sensitive function of the strength of the Hund's coupling, being very large for  $J_{Hund} = 0$  and unobservably small for  $J_{Hund} = 0.7$  in the temperature range considered. This is one of the main messages of this article.

To shed light into the coherence-incoherence crossover, we computed the resistivity due to electron-electron interactions, given by

$$\frac{1}{\rho} = \frac{\pi e^2}{V_0 \hbar} \int d\omega \left( -\frac{df}{d\omega} \right) \sum_{\mathbf{k}} \text{Tr}[v_{\mathbf{k}}(\omega) \rho_{\mathbf{k}}(\omega) v_{\mathbf{k}}(\omega) \rho_{\mathbf{k}}(\omega)] \quad (2)$$

where  $v_{\mathbf{k}}$  are velocities of electrons,  $\rho_{\mathbf{k}}$  is the spectral density of the electrons, and  $V_0$  is the volume of the unit cell. The resistivity in Fig. 2b demonstrates the crossover even clearly. The resistivity is approximately linear with modest slope above the coherence crossover temperature, and it exhibits a steep drop around the coherence temperature. It is important to stress that the drop of resistivity here is not due to proximity to a quantum critical point [9], or due to proximity to spin density wave [5], but because the electrons, being rather localized at high temperature, start to form a coherent quasiparticle bands with the underlying Fermi surface.

In most compounds, the strength of the correlations is controlled by the ratio of the diagonal on-site Coulomb interaction  $U$  to the bandwidth, while Hund's coupling usually plays a sub-leading role. We find that in this compound, a crucial role is played by the Hund's coupling. This point was first noticed in the context of a two band Hubbard model calculation at half-filling [21], and it was shown that the coherence scale is exponentially suppressed when Hund's coupling is taken into account. Moreover, it was emphasized that the non-rotationally invariant Hund's coupling, usually assumed in combination with traditional quantum Monte Carlo methods [22], can lead to a qualitatively and quantitatively wrong results. Indeed, we have found that the absence of the rotational invariant Hund's coupling leads to a first order phase transition as a function of  $J_{Hund}$ , between a Fermi liquid and non-Fermi liquid state at zero temperature. On the other hand, the rotational invariance of the Coulomb interaction

recovers Fermi liquid at zero temperature, although the coherence scale can become very small, as seen in Fig. 2b for  $J_{Hund} = 0.7$ .

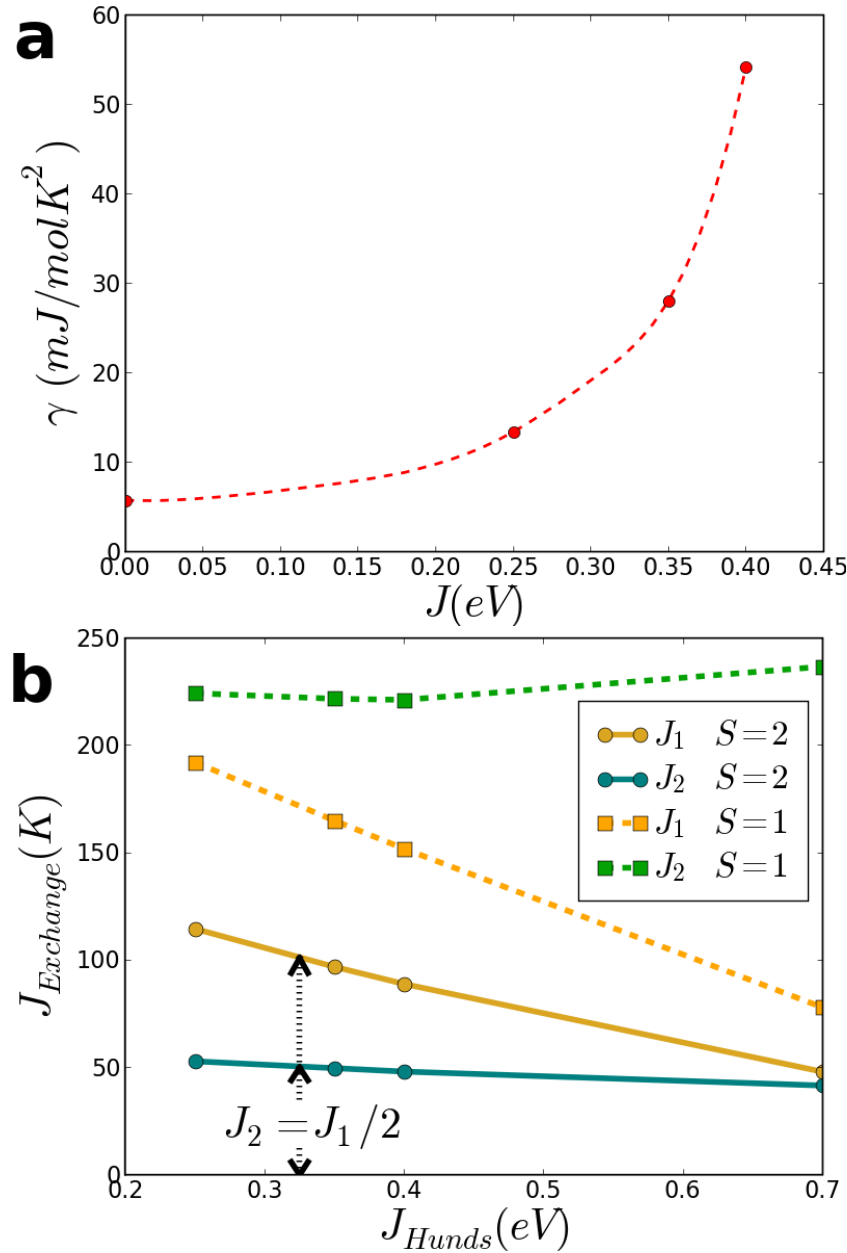
It is interesting to note that the atomic  $J_{Hund}$  in iron is approximately  $\sim 1.2$  eV. In many materials with correlated  $f$  orbitals, the screening of  $J_{Hund}$  in solid is well accounted for by 20% reduction from its atomic value. In LaOFeAs compound, only 30% of the atomic Hund's coupling gives the best agreement with experiment for resistivity and susceptibility.

To elucidate the sensitivity of mass enhancement with varying Hund's coupling, we show low temperature specific heat in Fig. 3. For small  $J_{Hund}$ , we find specific heat value very close to the LDA prediction  $\gamma \sim 6 mJ/(molK^2)$ . For  $J_{Hund} = 0.35$  eV, a value which gives resistivity in very favorable agreement with experiments, requires the enhancement of the specific heat for factor of 5 to  $30 mJ/(molK^2)$ . This is large enhancement and should be easy to measurable in future experiments, and is a key prediction of the theory. This would be a strong test that a local theory, as the one described in this paper, correctly predicts the physical properties of this material, since would allow to differentiate it from paramagnon or itinerant spin density wave theories, where the enhancement of the static susceptibility is much larger than the enhancement of the specific heat. At this point, reliable data for the normal state specific heat of the doped compound is not available down to sufficiently low temperature to reliably extract the coefficient  $\gamma$ . It would be interesting to measure it in the slightly doped compound (5%), where the superconducting temperature is low enough, and susceptibility shows the strongest enhancement.

The full computation of the momentum dependent spin response within LDA+DMFT is still a formidable task. However one can gain insights into the momentum dependence of  $\chi$  by making a simplifying assumptions, that near the Mott transition  $\chi(\mathbf{q})^{-1} = a + J_{\mathbf{q}}$ , where  $J_{\mathbf{q}}$  is the Fourier transform of the exchange constants, and  $a$  is a local but frequency dependent object.

We computed the first neighbor  $J_1$  and next-nearest neighbor  $J_2$  exchange constants as a function of Hund's coupling. For this purpose, we first constructed Wannier orbitals from iron  $3d$  bands (see appendix Appendix A), we exactly diagonalized the iron atom, and perform the second order perturbation theory, with respect to iron-iron hopping. This perturbation is carried out around the atomic ground state of the  $S = 2$  and  $S = 1$  sector.

Fig. 3b shows the exchange constants as a function of the Hund's rule coupling. This provides complementary information to the exchange constants computed around the band limit, which were recently determined by LDA [24, 25, 26], and are typically larger in magnitude than those obtained here. The exchange constants are supportive of the actual ordering in the parent compound, determined recently by the neutron scattering experiment[7], if  $J_2 > J_1/2$ . For reasonable Hund's coupling, both  $S = 1$  and  $S = 2$  ground states respect this inequality. If we select the  $S = 1$  state for the ground state, the next-nearest  $J_2$  is much bigger than  $J_1$ . For the actual ground state of the atom ( $S = 2$ ), and realistic  $J_{Hund} \sim 0.35$ , the value of  $J_2$  is close to



**Figure 3.** a) Low temperature Fermi liquid specific heat coefficient  $\gamma$  versus Hund's coupling strength. The effective mass is exponentially enhanced by the Hund's coupling. The coherence scale is consequentially exponentially suppressed by the Hund's coupling. b) Exchange constants, as obtained from the second order perturbation theory around the atomic limit, considering either  $S=2, N=6$  or  $S=1, N=6$  for the ground state. Positive values for the exchange constants correspond to antiferromagnetic couplings. The rest of the exchange constants are small, for example,  $J_3 \sim 1.8 \text{ K}$  and  $J_z \sim 0.2 \text{ K}$ .

$J_2 \geq J_1/2$ . It is interesting to note that according to a well known result [28], this particular choice of exchange constants results in large magnetic fluctuations due to frustration. At temperature above the spin density wave transition, there can also be a second order Ising phase transition, at which the two sublattice magnetizations of the



two interpenetrating Neel sublattices, lock together. According to Ref. [28], the Ising transition would occur around  $T_c \sim J_2 S^2 \sim 200K$ .

The large value of the next nearest neighbor exchange constant demonstrates that even in the atomic limit, the system is highly frustrated, as pointed out in Ref. [24, 27], which gives us more confidence that the single site DMFT description, employed in this article, is adequate, and the momentum dependent self-energy, as obtained by the cluster extensions of DMFT [17], will not qualitatively change the physics.

From the above provided evidence,  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$  system fits nicely within the general rubric of strongly correlated superconductivity. The hallmark of this phenomena, is the appearance of a incoherent normal state, above  $T_c$ . In the classic Bardeen-Cooper-Schrieffer theory of superconductivity, the effective Fermi energy is much larger than the pairing interaction, resulting in small critical temperatures. Strong correlations play the role of reducing the the kinetic energy, allowing the pairing interactions to lead to appreciable critical temperatures [29].

While these are common themes shared by all the high  $T_c$  materials, the mechanisms by which different systems manage to renormalize down their kinetic energies, and the details of the competing states that appear as a result, are different in different classes of materials.

Hole doped cuprate superconductors can be modeled as a single-band materials, with Coulomb correlation slightly above the Mott transition (in DMFT terminology the effective  $U$ , which arises from the charge transfer energy, is above  $U_{c2}$ ), and indeed maximal  $T_c$  is achieved when  $U$  is in the vicinity of critical  $U_{c2}$ .

We have argued in our earlier publication [12], that the  $\text{LaOFeAs}$  is an intrinsically multiorbital system, which is slightly *below*  $U_{c2}$ . The mechanism which is responsible for the incoherence of the normal state and the renormalization of the kinetic energy is the Hund's rule  $J_{\text{Hund}}$ , which we estimate here to be  $J_{\text{Hund}} = 0.35 - 0.4$  eV. Competing states in this material are magnetic states, that have been predicted in Ref. [5] and since observed by neutron scattering experiments [7, 6, 3].

The incoherent normal states, is then eliminated by either a magnetic or a superconducting state, and the study of the competition between these mechanism in the multiorbital framework, deserves further attention.

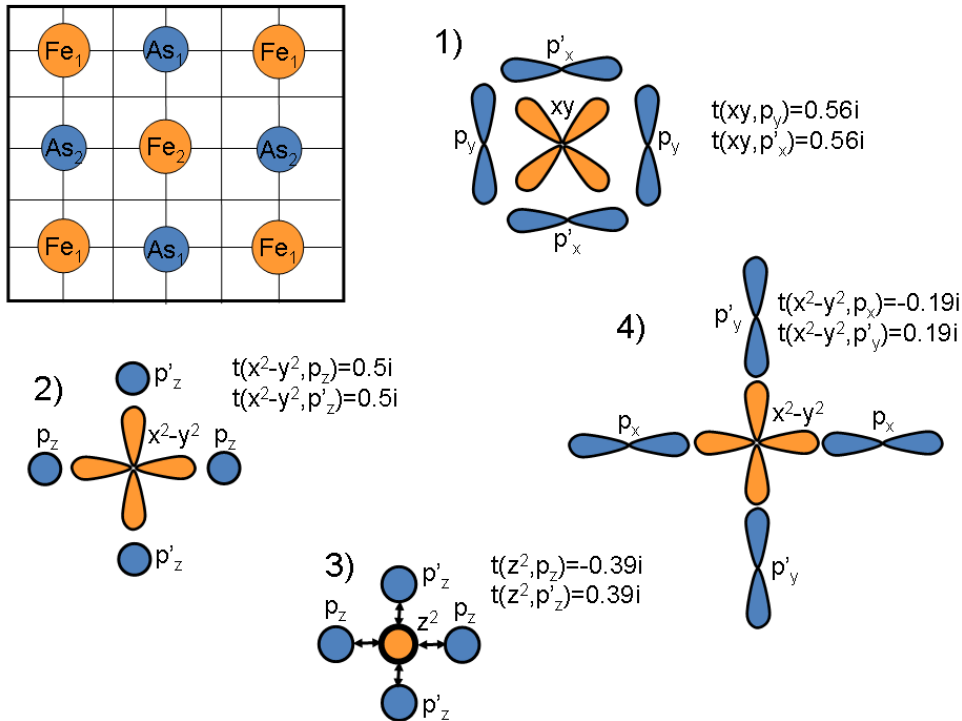
## **Appendix A. The tight binding Hamiltonian for the Fe-As planes**

The Fe-As planes in  $\text{LaFeAsO}$  can be described by a tight-binding Hamiltonian containing Fe and As atoms, with two Fe and two As per unit cell (see Fig. A1 for drawing of the unit cell containing  $\text{Fe}_1$ ,  $\text{Fe}_2$ ,  $\text{As}_1$  and  $\text{As}_2$ ). We obtained the tight-binding parameterization by downfolding the Local Density Approximation first-principles results, which are expressed in a well localized Muffin Tin Orbital (MTO) base. The localized nature of the MTO base makes the projection of Kohn-Sham orbitals to a localized orbital basis set simple, and downfolding procedure more constrained and well defined.

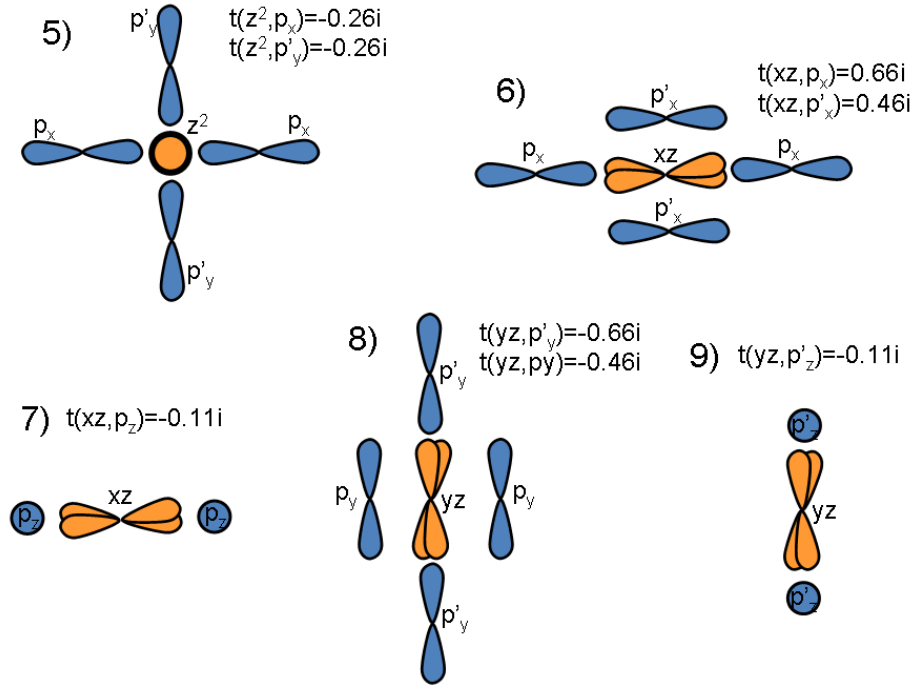
$E - \text{onsite}$	LaFeAsO	LaFePO
$E[x^2 - y^2]$	-0.510	-0.654
$E[yz]$	-0.535	-0.656
$E[z^2]$	-0.564	-0.635
$E[xy]$	-0.650	-0.692
$E[p_z]$	-2.144	-1.990
$E[p_x]$	-2.440	-2.358

**Table A1.** On-site energies for both LaFeAsO and LaFePO in the model with both Fe-d and As-p orbitals.

The few largest hoppings are sketched in Fig. A1 and A2. All energies and hoppings are in units of eV. The on-site energies of all orbitals are tabulated in table A1 and most of the hoppings are tabulated in tables A2 and A3. Note that orbitals are expressed in the coordinate system of the four atoms unit cell, shown in Fig. A1. The complete set of tight binding hoppings is available to download [23]. For comparison we also include the hoppings for a closely related compound LaFePO, which shows considerably less correlation effects due to somewhat larger crystal field splittings, which renormalize down the effective Hund's coupling.



**Figure A1.** left top) choice of the unit cell in the FeAs plane. As<sub>1</sub> lays above the Fe plane, and As<sub>2</sub> below it. 1-4) Sketch of some largest hoppings and their values.



**Figure A2.** Sketch of a few more relevant Fe-As hoppings.

Further downfolding to the 5 band model of Fe is possible. The on-site energies and the few largest hoppings of the corresponding multiorbital Hubbard model are tabulated in table 3. The sketch of the most relevant hoppings is shown in Fig. A3. Note that the coordinate system here is chosen differently than above. Here we use the  $45^\circ$  rotated coordinate system, which corresponds to usual square lattice of irons.

hopping	figure	LaFeAsO	LaFePO
$xy - p_y$	(1)	0.56*i	0.64*i
$xy - p'_x$	(1)	0.56*i	0.64*i
$x^2 - pz$	(2)	0.5*i	0.58*i
$x^2 - p'_z$	(2)	0.5*i	0.58*i
$x^2, p_x$	(4)	-0.19*i	-0.33*i
$x^2, p'_y$	(4)	0.19*i	0.33*i
$z^2, p_z$	(3)	-0.39*i	-0.45*i
$z^2, p'_z$	(3)	0.39*i	-0.45*i
$z^2, p_x$	(5)	-0.26*i	-0.22*i
$z^2, p'_y$	(5)	-0.26*i	-0.22*i
$xz, p_x$	(6)	0.66*i	0.76*i
$xz, p'_x$	(6)	0.46*i	0.51*i
$xz, p_z$	(7)	-0.11*i	0.0
$yz, p_y$	(8)	-0.46*i	-0.51*i
$yz, p'_y$	(8)	-0.66*i	-0.77*i
$yz, p'_z$	(9)	-0.11*i	0.0

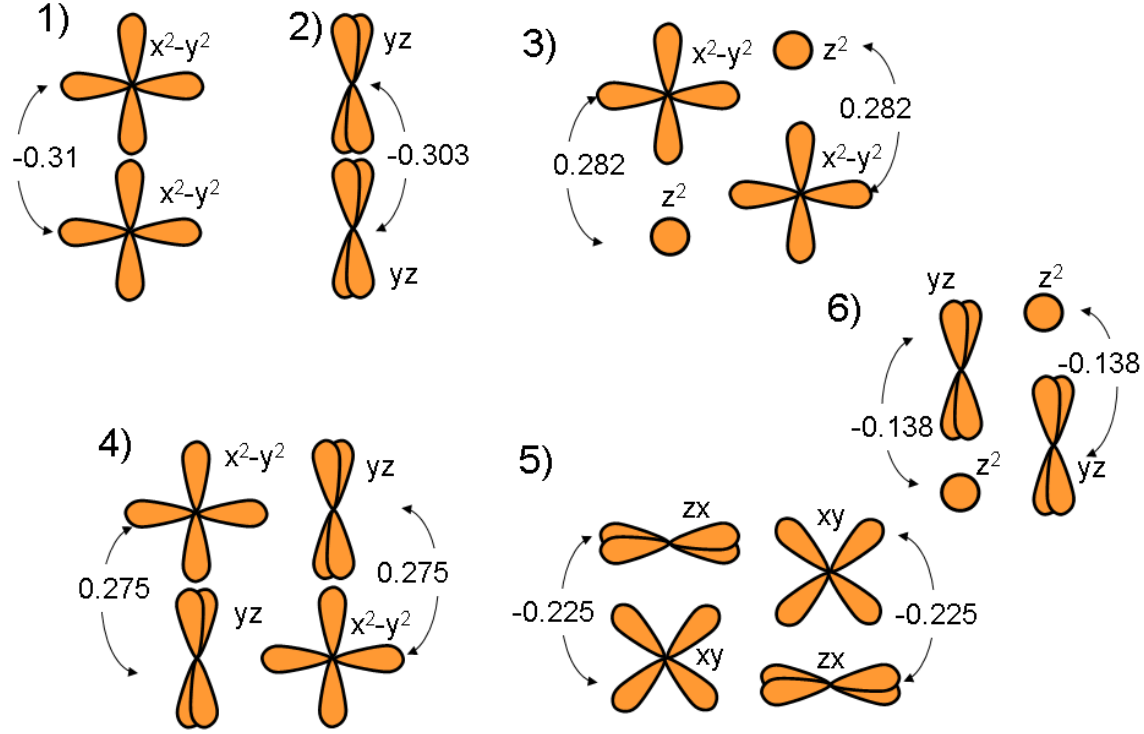
**Table A2.** The few largest hopping matrix elements in the model of Fe-d and As-p orbitals. Each Fe atom is surrounded by two inequivalent As-atoms. The two different As orbitals are sketched in Fig. A1,A2 and are denoted by  $p$  and  $p'$ .

- [1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, *Iron-Based Layered Superconductor*  $La[O_{1-x}F_x]FeAs$  ( $x=0.05-0.12$ ) with  $T_c=26K$ , J. Am. Chem. Soc., **130**, 3296 (2008).
- [2] Zhi-An Ren *et al.*, *Superconductivity at 55 K in iron-based F-doped layered quaternary compound*  $Sm[O_{1-x}F_x]FeAs$ , Chin. Phys. Lett. **25**, 2215 (2008).
- [3] Takatoshi Nomura *et al.*, *Crystallographic Phase Transition and High- $T_c$  Superconductivity in*  $LaOFeAs:F$ , Supercond. Sci. Technol. **21**, 125028 (2008).
- [4] D.J. Singh, M.H. Du, *LaFeAsO $_{1-x}F_x$ : A low carrier density superconductor near itinerant magnetism*, Phys. Rev. Lett. **100**, 237003 (2008).
- [5] J. Dong *et al.*, *Competing Orders and Spin-Density-Wave Instability in*  $La(O_{1-x}F_x)FeAs$ , Europhys. Lett., **83**, 27006 (2008).
- [6] M. A. McGuire *et al.*, *Phase transitions in LaFeAsO: Structural, magnetic, elastic, and transport properties, heat capacity and Mossbauer spectra*, Phys. Rev. B **78**, 094517 (2008).
- [7] Clarina de la Cruz *et al.*, *Magnetic Order versus superconductivity in the Iron-based layered*  $La(O_{1-x}F_x)FeAs$  systems, Nature **453**, 899 (2008).
- [8] L. Ding *et al.*, *Specific heat of iron-based high- $T_c$  superconductor*  $SmO_{1-x}F_xFeAs$ , Phys. Rev. B **77**, 180510(R) (2008).
- [9] R. H. Liu *et al.*, *Phase Diagram and Quantum Critical Point in Newly Discovered Superconductors:*  $SmO_{1-x}F_xFeAs$ , Phys. Rev. Lett. **101**, 087001(2008).
- [10] Athena S. Sefat *et al.*, *Electron Correlations in the Low Carrier Density*  $LaFeAsO_{0.89}F_{0.11}$  Superconductor ( $T_c = 28$  K), Phys. Rev. B **77**, 174503 (2008).
- [11] Xiyu Zhu *et al.*, *Upper critical field, Hall effect and magnetoresistance in the iron-based layered superconductor*  $LaO_{0.9}F_{0.1-\delta}FeAs$ , Supercond. Sci. Technol. **21**, 105001 (2008).
- [12] K. Haule, J. H. Shim, G. Kotliar, *Correlated electronic structure of LaOFeAs*, Phys. Rev. Lett. **100**, 226402 (2008).

hopping	LaFeAsO	LaFePO
$p_z, p'_z$	0.29	0.27
$p_x, p'_x$	0.16	0.21
$p_y, p'_y$	0.16	0.21
$p_x, p'_z$	-0.25	-0.27
$p_y, p'_z$	0.25	0.27
$p_z, p'_x$	-0.25	-0.27
$p_z, p'_y$	0.25	0.27
$p_x, p'_y$	-0.15	-0.18
$p_y, p'_x$	-0.15	-0.18
$xy, xy'$	-0.30	-0.35
$xy, z^{2'}$	0.21	0.24
$z^2, xy'$	0.21	0.24
$x^2, x^{2'}$	0.19	0.18
$yz, xz'$	0.15	0.16
$xz, yz'$	0.15	0.16
$yz, x^{2'}$	0.09	0.12
$xz, x^{2'}$	0.09	0.12
$x^2, yz'$	0.09	0.12
$x^2, xz'$	0.09	0.12
$z^2, z^{2'}$	-0.07	-0.06
$yz, yz'$	0.06	0.07
$yz, xy'$	0.06	0.07
$xz, xz'$	0.06	0.07
$xz, xy'$	-0.06	-0.07
$xy, yz'$	0.06	0.07
$xy, xz'$	-0.06	-0.07
$yz, z^{2'}$	-0.02	-0.02
$xz, z^{2'}$	0.02	0.02
$z^2, yz'$	-0.02	-0.02
$z^2, xz'$	0.02	0.02

**Table A3.** Continued from table A2, a few more large hopping matrix elements of the Fe-d,As-p model.

- [13] L. Boeri, O. V. Dolgov, A. A. Golubov, *Is  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$  an electron-phonon superconductor ?*, Phys. Rev. Lett. **101**, 026403 (2008).
- [14] Kazuhiko Kuroki *et al.*, *Unconventional superconductivity originating from disconnected Fermi surfaces in  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$* , Phys. Rev. Lett. **101**, 087004 (2008).
- [15] Zi-Jian Yao *et al.*, *Spin Fluctuations, Interband Coupling, and Unconventional Pairing in Iron-based Superconductors*, arXiv:0804.4166 (unpublished).
- [16] Xiao-Liang Qi *et al.*, *Pairing strengths for a two orbital model of the Fe-pnictides*, arXiv:0804.4332 (unpublished).



**Figure A3.** The sketch of the largest Fe-Fe hoppings in the effective five band Hubbard model of irons only.

- [17] G. Kotliar *et al.*, *Electronic structure calculations with dynamical mean-field theory*, Rev. Mod. Phys. **78**, 865 (2006).
- [18] K. Haule, *Quantum Monte Carlo Impurity Solver for Cluster DMFT and Electronic Structure Calculations in Adjustable Base*, Phys. Rev. B **75**, 155113 (2007).
- [19] Technically, the critical value of the Hubbard  $U$ , which is needed to obtain a insulating state ( $U_{c2}$ ) in an SU( $N$ ) symmetric model is known to scale linearly with orbital degeneracy as  $N$ . For this reason, Ref. [22]v1, which ignored the Hund's rule coupling, concluded that these materials are non correlated metals.
- [20] K. Haule, *Quantum Monte Carlo Impurity Solver for Cluster DMFT and Electronic Structure Calculations in Adjustable Base*, Phys. Rev. B. **75**, 155113 (2007).
- [21] Th. Pruschke and R. Bulla, *Hund's coupling and the metal-insulator transition in the two-band Hubbard model*, Eur. Phys. J. B **44**, 217 (2005).
- [22] A.O. Shorikov *et al.*, *Coulomb correlation effects in LaOFeAs: LDA+DMFT(QMC) study*, arXiv:0804.3283v1 (unpublished).
- [23] The full Hamiltonian for 5-band Fe model and 8-band Fe-As model can be downloaded from:  
`\protect\vrule width0pt\protect\href{http://www.physics.rutgers.edu/~haule/FeAs/}{http://www`
- [24] T. Yildirimar, *Origin of the 150K Anomaly in LaOFeAs; Competing Antiferromagnetic Superexchange Interactions, Frustration, and Structural Phase Transition*, Phys. Rev. Lett. **101**, 057010 (2008).
- [25] Fengjie Ma *et al.*, *Antiferromagnetic superexchange interactions in LaOFeAs*, arXiv:0804.3370 (unpublished).

hops	fig	LaFeAsO	LaFePO
$E[xy]$		0.096	0.244
$E[yz]$		-0.034	-0.054
$E[z^2]$		-0.189	-0.201
$E[x^2]$		-0.251	-0.162
$x^2, x^{2'}$	(1)	-0.310	-0.343
$yz, yz'$	(2)	-0.303	-0.356
$x^2, z^{2'}$	(3)	0.282	0.310
$z^2, x^{2'}$	(3)	0.282	0.310
$x^2, yz'$	(4)	0.275	0.356
$yz, x^{2'}$	(4)	0.275	0.356
$zx, xy'$	(5)	-0.225	-0.269
$xy, zx'$	(5)	-0.225	-0.269
$yz, z^{2'}$	(6)	-0.138	-0.104
$z^2, yz'$	(6)	-0.138	-0.104
$zx, zx'$		-0.010	-0.035
$xy, xy'$		-0.086	-0.175
$z^2, z^{2'}$		0.036	0.088
$zx, z^{2'}$		0.004	0.003
$z^2, zx'$		0.004	0.003
$zx, x^{2'}$		-0.003	0.006
$x^2, zx'$		0.003	-0.006

**Table A4.** The on-site energies and the most important hoppings in the five band model of Fe-d orbitals only.

- [26] Z. P. Yin *et al.*, *Electron-Hole Symmetry and Magnetic Coupling in Antiferromagnetic LaOFeAs*, Phys. Rev. Lett. **101**, 047001 (2008).
- [27] Qimiao Si, Elihu Abrahams, *Strong Correlations and Magnetic Frustration in the High Tc Iron Pnictides*, Phys. Rev. Lett. **101**, 076401 (2008).
- [28] P. Chandra, P. Coleman, and A.I. Larkin, *Ising transition in frustrated Heisenberg models*, Phys. Rev. Lett. **64**, 88 (1990).
- [29] M. Capone, M. Fabrizio, C. Castellani, E. Tosatti, *Strongly Correlated Superconductivity*, Science **296**, 2364 (2002).